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Induced Anisotropy in FeCo-Based Nanocrystalline Ferromagnetic Alloys (HITPERM) by Very High Field Annealing

F. Johnson, H. Garmestani, S. Y. Chu, M. E. McHenry, and D. E. Laughlin

Abstract—Very high magnetic field annealing is shown to affect the magnetic anisotropy in FeCo-base nanocrystalline soft ferromagnetic alloys. Alloys of composition $\text{Fe}_{44.5}\text{Co}_{44.5}\text{Zr}_7\text{B}_4$ were prepared by melt spinning into amorphous ribbons, then wound to form toroidal bobbin cores. One set of cores was crystallized in a zero field at 600 °C for 1 h, then, field annealed at 17 tesla (T) at 480 °C for 1 h. Another set was crystallized in a 17-T field at 480 °C for 1 h. Field orientation was transverse to the magnetic path of the toroidal cores. An induced anisotropy is indicated by a sheared hysteresis loop. Sensitive torque magnetometry measurements with a Si cantilever sensor indicated a strong, uniaxial, longitudinal easy axis in the zero-field-crystallized sample. The source is most likely magnetoelastic anisotropy, caused by the residual stress from nanocrystallization and the nonzero magnetostriction coefficient for this material. The magnetostrictive coefficient λ_s is measured to be 36 ppm by a strain gage technique. Field annealing reduces the magnitude of the induced anisotropy. Core loss measurements were made in the zero-field-crystallized, zero-field-crystallized-than-field-annealed, and field-crystallized states. Core loss is reduced 30%–50% (depending on frequency) by field annealing. X-ray diffraction reveals no evidence of crystalline texture or orientation that would cause the induced anisotropy. Diffusional pair ordering is thought to be the cause of the induced anisotropy. However, reannealing the samples in the absence of a magnetic field at 480 °C does not completely remove the induced anisotropy.

Index Terms—FeCo alloys, field annealing, magnetic anisotropy, nanocrystals, soft magnetic alloys.

I. INTRODUCTION

HITPERM [1] is a nanocomposite soft ferromagnetic alloy comprised of $\alpha(\alpha')$ -FeCo nanocrystalline grains embedded in a residual amorphous matrix. HITPERM is distinguished from other nanocomposite alloys (such as FINEMET [2] and NANOPERM [3]) by a high Curie temperature (T_c) of both the nanocrystalline and amorphous phases [4]. This

enables the grains to remain exchange coupled to high temperatures.

The soft magnetic properties of nanocomposite alloys derive from the exchange averaging of magnetocrystalline anisotropy of the nanocrystalline grains, as described with the random anisotropy model (RAM) [5]. The observed coercivity (H_c) of HITPERM is larger than predicted by the RAM for HITPERM's measured grain size. Extensions to the RAM can account for this, but the simplest explanation can be the presence of a uniaxial magnetoelastic anisotropy. Beyond H_c , the ac power loss (P_c) and relative permeability (μ_r) are important figures of merit for soft ferromagnets. The contribution to P_c that will be considered here is the hysteretic loss. The magnitude of the hysteretic loss is determined by the relative proportion of irreversible (domain wall motion) and reversible (rotation) magnetization processes.

Field annealing is an established technique for inducing a magnetic anisotropy in a chosen direction in both crystalline and amorphous materials [6]. If the induced anisotropy is transverse to the magnetic path of the sample, then reversible rotational magnetization processes are activated. Thus, field annealing can provide a mechanism for reducing P_c . A tradeoff that must be made is that a transverse anisotropy reduces the permeability as well. A necessary precondition for field annealing is that the annealing temperature (T_a) must be below the T_c of the material. The operative mechanism has been postulated to be pair ordering of unlike atoms aligned with the applied field [6]. This is a diffusional process, and may be aided by the presence of free volume in an amorphous phase. A structural source (crystalline texture/defects, granular shape morphology) may contribute to the anisotropy if it can be shown to evolve in response to the applied field.

FINEMET is commercially available in the field-annealed state. Herzer [7] reports that the magnitude of the anisotropy is proportional to the metalloid content, with a maximum of 100 J/m³ at the composition $\text{Fe}_{73.5}\text{Cu}_1\text{Nb}_3\text{Si}_{13.5}\text{B}_9$. The mechanism of induced anisotropy is assumed to be Fe–Si pair ordering in the nanocrystalline phase. Lovas *et al.* [8] report a maximum field induced anisotropy for NANOPERM of 50 J/m³. The mechanism in NANOPERM is presumed to be Fe–Zr pair ordering in the nanocrystalline phase.

II. PROCEDURE

Toroidal cores of melt spun $\text{Fe}_{44.5}\text{Co}_{44.5}\text{Zr}_7\text{B}_4$ were produced at the Carnegie Mellon University. Transverse field

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F. Johnson is with the National Institute of Standards and Technology, Gaithersburg, MD 20899-8552 USA and also with the Materials Science and Engineering Department, Carnegie Mellon University, Pittsburgh, PA 15213 USA (e-mail: frank.johnson@nist.gov).

H. Garmestani is with the Materials Science and Engineering Department, Georgia Institute of Technology, Atlanta, GA 30332-0245, USA (e-mail: hamid.garmestani@mse.gatech.edu).

S.-Y. Chu, M. E. McHenry, and D. E. Laughlin are with the Materials Science and Engineering Department, Carnegie Mellon University, Pittsburgh, PA 15213 USA (e-mail: sc79@andrew.cmu.edu; mm7g@andrew.cmu.edu; dl0p@andrew.cmu.edu).

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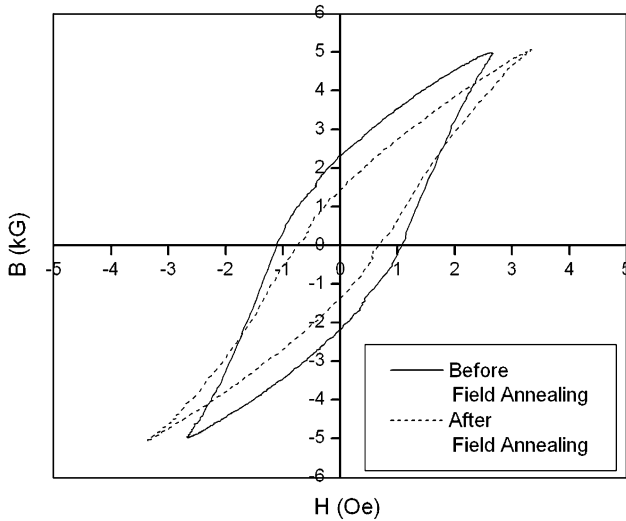


Fig. 1. Hysteresis loop at 5-kG BMax, 1 kHz of the same nanocrystalline HITPERM core before and after field annealing. Field annealing is observed to shear the loop, lowering permeability and decreasing coercivity.

annealing was performed at the National High Field Magnet Laboratory, Tallahassee, FL. Three processing conditions were studied. In the first, called *zero-field-crystallized*, samples were heated to 600 °C in the absence of a field for 1 h, then furnace cooled. In the second, called *zero-field-crystallized-than-field-annealed*, the samples were annealed in zero field at 600 °C for 1 h, furnace cooled, then annealed in a 17-T transverse field at 480°C for 1 h, then furnace cooled in a field. In the third, called *field crystallized*, initially amorphous samples were annealed in a 17-T transverse field at 480 °C for 1 h, then furnace cooled in a field. A subset of *zero-field-crystallized-than-field-annealed* and *field-crystallized* samples, were reannealed in the absence of a field at 480 °C for 1 h, then furnace cooled. All annealing was done in a flowing Ar atmosphere.

Core loss measurements were made with a Walker LDJ AMH-401A Hysteresisgraph. Torque magnetometry was performed at room temperature in a 5-T applied field by a Quantum Design Physical Properties Measurement System (PPMS) with a Si cantilever. Torque magnetometry samples were a stack of ten 1 mm × 1 mm square ribbons glued with epoxy. Magnetostriction measurements were made at room temperature with a strain gage technique using the resistance bridge of the PPMS at room temperature with a 1-T applied field. X-ray diffraction was made at normal and grazing incidence with a Phillips X'Pert Pro diffractometer equipped with a Cu tube and capillary X-ray lens.

III. RESULTS

HITPERM in the *zero-field-crystallized-than-field-annealed* state shows a hysteresis loop that is sheared relative to the *zero-field-crystallized* state as shown in Fig. 1. These loops were collected on the same sample, under the same conditions, the only variable being the presence or absence of field annealing. Field annealing HITPERM reduces core loss 30%–50% at low frequencies as shown in Fig. 2. The reduction in core loss is less

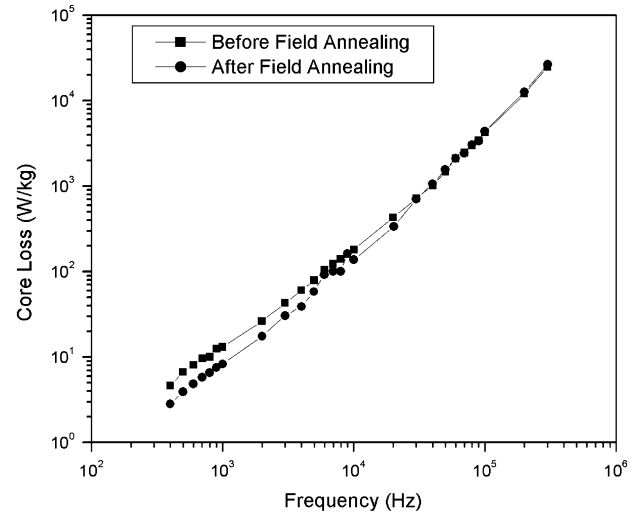


Fig. 2. Core loss versus frequency, at 5-kG BMax, of the same nanocrystalline HITPERM core before and after field-annealing. Core loss is observed to be reduced up to 50% at low frequencies. Effect of field annealing is reduced at high frequencies due to dominance of eddy current losses.

TABLE I
UNIAXIAL ANISOTROPY MEASURED BY TORQUE MAGNETOMETRY

| Sample Condition | Anisotropy energy density (J/m ³) | Angular position of easy axis relative to torque cantilever stage zero (°) |
|--|---|--|
| <i>zero-field-crystallized</i> | 119,000 | 4 |
| <i>zero-field-crystallized-than-field-annealed</i> | 57,000 | 1 |
| <i>field-crystallized</i> | 28,000 | 15 |

pronounced at high frequencies due to the larger contribution of eddy current losses.

Torque magnetometry directly measured the magnetic anisotropy. All samples displayed uniaxial anisotropy. The anisotropy energy densities and angular position of the easy axes (relative to the zero position of the torque cantilever stage) are given in Table I. The position of the easy axes are all close to longitudinal. The samples were positioned on the cantilever stage by hand and thus the small shifts in the easy axes cannot be conclusively attributed to the field annealing. However, the anisotropy energy density is significant and does follow an expected trend: *zero-field-crystallized* exhibits the highest, and the *field-crystallized* state is the lowest. Conventional and grazing-incidence X-Ray diffraction patterns were collected for all three annealing conditions. No difference in the relative heights of the peaks was detected in either configuration between any of the annealing conditions.

A subset of the *zero-field-crystallized-than-field-annealed* and *field-crystallized* samples were reannealed at 480 °C in zero field for 1 h in an attempt to remove the induced anisotropy. Table II presents the change in core loss after reannealing. In all cases core loss was observed to increase after reannealing. However, in the case of the *zero-field-crystallized-than-field-annealed* samples the core loss was not observed to return to the *zero-field-crystallized* state.

The λ_S value of nanocrystalline HITPERM was observed to be 36 ppm. This is in general agreement with similar composi-

TABLE II
CORELOSS (MILLIWATTS/GRAM) AS FUNCTION OF SAMPLE CONDITION AND
BMAX, MEASURED AT 1 KHz

| Sample condition | BMax (kG) | | |
|---|-----------|-----|----|
| | 1 | 5 | 10 |
| <i>zero-field-crystallized</i> | .90 | 13 | 26 |
| <i>zero-field-crystallized- than-field-annealed</i> | .38 | 9.0 | 18 |
| <i>reannealed</i> | .65 | 10 | 21 |

tions of nanocrystalline material [9]. The strain gage technique is expected to have an accuracy of 20% [10].

IV. DISCUSSION

The uniaxial anisotropy that is present in *zero-field-crystallized* HITPERM is quite large in comparison to other soft magnetic materials. For example, the first magnetocrystalline anisotropy constant for cubic Fe is 4.8×10^4 J/m³, for uniaxial Co is 4.1×10^5 J/m³, and for disordered cubic FeCo is -1.5×10^4 J/m³ [11]. The anisotropy energy density in nanocrystalline HITPERM is comparable to Co. No cubic or higher order components of the torque curve were detected, indicating that magnetocrystalline anisotropy was not a major contribution. Shape anisotropy was not a contribution, since the applied field fully saturated the sample. Thus, the anisotropy was most likely caused by the magnetoelastic anisotropy due to coupling of the magnetostriction with residual stress in the nanocrystalline structure.

The magnetoelastic energy density is given by $(3/2)\lambda_s\sigma_i$. The residual stress is thus calculated to be 2.2 GPa. It is not possible that the epoxy mounting of the samples could provide this stress due to the epoxy's low shear strength (10 MPa). Bozorth [12] derived a formula for calculating the residual stress from the initial permeability μ_o

$$\mu_o - 1 = \frac{8\pi B_S^2}{9\lambda_s\sigma_i}. \quad (1)$$

The relative μ_o for *zero-field-crystallized* HITPERM was measured to be 650 at 0.4 A/m maximum H at 1 kHz. For a B_S of 1.6 T, this yields a residual stress of 3 GPa. Fe-metalloid metallic glasses have been reported to have a Young's modulus on the order of 160 GPa [13]. A residual stress of 2.2 GPa would result from a volume contraction of 4% during crystallization. This is larger than expected for amorphous to crystalline volume change. Given the accuracy of the magnetostriction measurement and the unknown modulus of the amorphous phase in HITPERM, a residual stress on the order of 2–3 GPa is reasonable. An anisotropic distribution of residual stress would be required to create such a magnetoelastic anisotropy.

The induction of anisotropy by field annealing is evident in both the core loss and torque data. The magnitude of the reduction at low frequencies appears to be similar to the magnitude of the reduction of longitudinal anisotropy energy density. The

field induced anisotropy in HITPERM is much larger than that observed in FINEMET or NANOPERM, suggesting a mechanism other than pair-ordering. The presence of crystalline texture would invalidate one premise of the RAM: that the spatial distribution of easy axes be random. However, X-ray diffraction did not indicate such a texture. Reannealing the samples in the absence of a field did not completely remove the induced anisotropy, indicating that a structural source other than diffusional pair ordering may be present (it must be noted that the presence of a field will change activation energies for diffusion).

V. CONCLUSION

Transverse field annealing was successful in changing the magnetic anisotropy in crystallized samples of HITPERM. Field annealing and field crystallization reduced the measured uniaxial anisotropy energy densities. Core loss was lowered by up to 50% at low frequencies. No evidence of crystalline texture was found. The induced anisotropy was not completely erased by reannealing. A large longitudinal, uniaxial anisotropy was found in all samples, most likely due to magnetoelastic anisotropy.

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